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(54) TACKINESS IMPARTING AGENT FOR HOT MELT SEALING MATERIAL AND HOT MELT SEALING MATERIAL

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a hot melt sealing material having excellent light fastness and a tackiness imparting agent for the hot melt sealing material by including a thermoplastic elastomer as a base polymer.

SOLUTION: This tackiness imparting agent for a hot melt sealing material is composed of a hydrogenated aromatic hydrocarbon resin obtained by polymerizing a polymerizable monomer in a 9C fraction and its polymerizable monomer contains ≥ 50 wt.% of a vinyltoluene and ≤ 20 wt.% of indene, and a degree of hydrogenation of an aromatic ring in the aromatic hydrocarbon resin is $\geq 50\%$.

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CLAIMS

[Claim(s)]

[Claim 1] The tackifier for hot melt sealing materials which it is the tackifier for hot melt sealing materials which comes to use the hydride of the aromatic series system hydrocarbon resin obtained by carrying out the polymerization of the polymerization nature monomer in C9 fraction, and this polymerization nature monomer contains vinyltoluene 50% of the weight or more, and contains an indene at 20 or less % of the weight of a rate, and is characterized by the rate of hydrogenation of the ring of aromatic series system hydrocarbon resin being 50% or more.

[Claim 2] The tackifier for hot melt sealing materials according to claim 1 whose ring and ball softening point of the hydride of aromatic series system hydrocarbon resin is 60-130 degrees C.

[Claim 3] thermoplastic elastomer -- (-- A --) -- containing -- becoming -- the base -- a polymer -- 100 -- weight -- the section -- receiving -- being according to claim 1 or 2 -- hot melt -- a sealing material -- ** -- a tackifier -- (-- B --) -- 20 - 150 -- weight -- the section -- and -- a plasticizer -- (-- C --) -- five - 100 -- weight -- the section -- blending -- becoming -- hot melt -- C -- RUNGU -- material .

[Claim 4] The hot melt sealing material according to claim 3 whose thermoplastic elastomer (A) is the hydride and/or its denaturation object of a styrene-conjugated diene system block copolymer.

[Claim 5] Hot melt C RUNGU material according to claim 3 or 4 whose content of thermoplastic elastomer (A) in hot melt C RUNGU material is 15 - 70 % of the weight.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the tackifier for hot melt sealing materials, and a hot melt sealing material. The hot melt sealing material of this invention can use for junction to various kinds of base materials, such as junction of window frames, such as junction on the lens of for example, the tail lamp for cars, or a head lamp, and the housing body, a house, and an automatic vending machine, or junction of the lamp of a signal, and a base material.

[0002]

[Description of the Prior Art] Conventionally, various kinds of sealing materials which make a base polymer thermosetting polyurethane, a polysulfide, silicone, or a thermoplastic polymer are used for junction to various base materials and a base material.

[0003] Since it excels in colorlessness and transparency, the sealing material which makes thermoplastic elastomer a base polymer also in said thermoplastic polymer is used as a clear sealing material of the part where importance is attached to colorlessness, such as junction on the lens and the housing body which are used for the tail lamp for cars, or a head lamp, and transparency. Moreover, since junction of a base material is performed by hot melt (thermofusion liquid) coating, compared with the sealing material of the type which joins completely the base material which makes a base polymer thermoplastic polymers, such as thermosetting polyurethane, and ethylene / vinyl acetate copolymer, a polyamide, exfoliation of a base material is comparatively easy for this clear sealing material, and also when performing the exchange and collection by type of a base material by type, he is desirable.

[0004] The clear sealing material which makes such thermoplastic elastomer a base polymer is indicated by for example, a U.S. Pat. No. 429008 specification, JP,6-313159,A, JP,7-169307,A, etc. Fundamentally, the clear sealing material indicated by these blends a tackifier, a plasticizer, etc. with thermoplastic elastomer, and raises an adhesive property etc., as a tackifier, it is colorlessness and the good hydrogenation petroleum resin of transparency is mainly used.

[0005] However, the clear sealing material containing tackifiers, such as hydrogenation petroleum resin, cannot say lightfastness with it being enough. namely, the optical exposure of the sunlight which attains to long duration -- a clear sealing material -- yellowing -- in order to color, it is easy to spoil an appearance, and the clear sealing material itself deteriorates depending on the case, and there is a trouble of spoiling the function as a sealing material itself. [0006]

[Problem(s) to be Solved by the Invention] This invention aims at offering the tackifier for hot melt sealing materials used for the hot melt sealing material which came to contain thermoplastic elastomer as a base polymer, and was excellent in lightfastness, and the hot melt sealing material concerned.

[0007]

[Means for Solving the Problem] this invention persons came to complete header this invention for the ability of said purpose to be attained by using the hydride of the specific aromatic series system hydrocarbon resin shown below as a tackifier for hot melt sealing materials, as a result of repeating examination wholeheartedly that said technical problem should be solved.

[0008] Namely, this invention is a tackifier for hot melt sealing materials which comes to use the

hydride of the aromatic series system hydrocarbon resin obtained by carrying out the polymerization of the polymerization nature monomer in C9 fraction. This polymerization nature monomer contains vinyltoluene and contains an indene at 20 or less % of the weight of a rate 50% of the weight or more, and to the tackifier for hot melt sealing materials and pan which are characterized by the rate of hydrogenation of the ring of aromatic series system hydrocarbon resin being 50% or more thermoplastic elastomer -- (A) -- containing -- becoming -- the base -- a polymer -- 100 -- weight -- the section -- receiving -- said -- hot melt -- a sealing material -- ** -- a tackifier -- (B) -- 20 - 150 -- weight -- the section -- and -- a plasticizer -- (C) -- five - 100 -- weight -- the section -- blending -- becoming -- hot melt -- C -- RUNGU -- material -- being related .

[0009]

[Embodiment of the Invention] The tackifier for hot melt sealing materials of this invention (B) is the hydride of the aromatic series system hydrocarbon resin obtained by carrying out the polymerization of the polymerization nature monomer in C9 fraction, this polymerization nature monomer contains vinyltoluene 50% of the weight or more, and contains an indene at 20 or less % of the weight of a rate, and the rate of hydrogenation of the ring of aromatic series system hydrocarbon resin is 50% or more of thing.

[0010] C9 fraction usually means what was obtained according to cracking of naphtha, and the compound of high-boiling in addition points, such as styrene, vinyltoluene, alpha methyl styrene, and indenenes, etc. is contained as a polymerization nature monomer in C9 fraction. C9 general fraction usually contains vinyltoluene and an indene about 40% of the weight, respectively, and about remaining 20 % of the weight consists of styrene etc. In this invention, by distilling such C9 fraction, the indenenes of C9 fraction, the compound of a high-boiling point, etc. are removed, and the vinyltoluene content in a polymerization nature monomer uses what was prepared so that the content of an indene might become 20 or less % of the weight at 50 % of the weight or more. It is 60 % of the weight or more more preferably 55% of the weight or more, it is so good that many, and preferably, the content of an indene is 10 or less % of the weight more preferably, and it is preferably good [few vinyltoluene contents] so that there are 15 or less % of the weight. When the content of the vinyltoluene in a polymerization nature monomer is less than 50%, or when the content of an indene exceeds 20 % of the weight, it becomes impossible to be satisfied with the hydride of the aromatic series system hydrocarbon resin obtained of lightfastness. In addition, aromatic series system hydrocarbon resin is obtained by carrying out the polymerization of this polymerization nature monomer by the usual cationic polymerization.
 [0011] Said aromatic series system hydrocarbon resin is hydrogenated so that the rate of hydrogenation of a ring may become 50% or more further. Lightfastness cannot be satisfied when the rate of hydrogenation of a nucleus is less than 50%.

[0012] A hydrogenation reaction is performed by adjusting conditions suitably to the bottom of existence of a hydrogenation catalyst so that the rate of hydrogenation of said aromatic series system hydrocarbon resin may become said within the limits (the rate of hydrogenation of a ring is 50% or more).

[0013] As a hydrogenation catalyst, various kinds of things, such as metallic compounds, such as metals, such as nickel, palladium, platinum, cobalt, a rhodium, a ruthenium, and molybdenum, or these oxides, and a sulfide, can be used. This hydrogenation catalyst may be supported and used for support, such as an alumina with big surface area, a silica (diatom earth), carbon, and a titania, by porosity. It is desirable to use a nickel-diatom earth catalyst also in these catalysts in this invention from that it is easy to adjust the rate of hydrogenation to said within the limits or a costs side. 0.1- of the aromatic series system hydrocarbon resin whose amount of the catalyst used is raw material resin -- it is 0.1 - 2 % of the weight preferably about 3% of the weight.

[0014] A hydrogenation pressure is performed in the about two 30 - 300 kg/cm range, and the conditions of a hydrogenation reaction usually perform reaction temperature in about 150-300 degrees C. A hydrogenation pressure is 100 - 200 kg/cm² preferably, and reaction temperature is 200-280 degrees C. When a hydrogenation pressure does not fulfill 30 kg/cm², or when reaction temperature does not fulfill 150 degrees C, hydrogenation cannot progress easily, and when a hydrogenation pressure exceeds 300 kg/cm², or when reaction temperature exceeds 300

degrees C, there is an inclination for decomposition to take place and for softening temperature to fall. Moreover, reaction time is usually 2 - 7 hours preferably for about 1 to 7 hours. Said hydrogenation reaction is performed in the condition of having fused aromatic series system hydrocarbon resin, or having dissolved in the solvent. A cyclohexane, n-hexane, n-heptane, a decalin, etc. can be used as a solvent.

[0015] In addition, although the case where a batch process was adopted as a reaction format about the amount used and reaction time of a catalyst was explained, as a reaction format, circulation types (a fixed-bed type, fluidized bed, etc.) are also employable.

[0016] In this way, as for about 60-130 degrees C and number average molecular weight, 300 to about 2000 are [the softening temperature of the hydride of the obtained aromatic series system hydrocarbon resin] usually desirable.

[0017] The hot melt sealing material of this invention blends said tackifier for hot melt sealing materials (B) and plasticizer (C) with the base polymer containing thermoplastic elastomer (A).

[0018] As thermoplastic elastomer (A), the copolymer or block copolymer of styrene, such as styrene and methyl styrene, and conjugated dienes, such as a butadiene and an isoprene, is raised. Although especially the ratio of the weight ratio of styrene/conjugated dienes is not restricted, it is usually about 10 / 90 to 50/50.

[0019] As thermoplastic elastomer (A) being concrete, a styrene-butadiene copolymer, a styrene-isoprene copolymer, a styrene-isoprene-styrene triblock copolymer (SIS), a styrene-butadiene-styrene triblock copolymer (SBS), etc. can be illustrated, for example. Furthermore, as thermoplastic elastomer (A), what hydrogenated said copolymer or block copolymers, such as a hydride of a styrene-butadiene copolymer, a hydride of a styrene-isoprene copolymer, a hydride [(styrene/ethylene) - (butylene/styrene) copolymer] (SEBS) of SBS, and a hydride [(styrene/ethylene) - (propylene/styrene) copolymer] (SEPS) of SIS, and its denaturation object are raised. In addition, various kinds of denaturation objects, such as what especially the rate of hydrogenation of said copolymer or a block copolymer was not **ed, and introduced polar groups, such as a carboxyl group and a hydroxyl group, into some of said copolymers or block copolymers as a denaturation object, are raised. As thermoplastic elastomer (A) of this this invention, the hydride and/or its denaturation objects of a styrene-conjugated diene system block copolymer, such as SEBS and SEPS, are desirable at the point of excelling in lightfastness.

[0020] Since the adhesion to adherend and the adhesive property at the time of heating are raised in addition to said thermoplastic elastomer (A), thermoplastic polymers other than said thermoplastic elastomer (A) can also be used for the base polymer of the hot melt sealing material of this invention. As such a thermoplastic polymer, ethylene / vinyl acetate copolymer (EVA), an ethylene ethyl acrylate copolymer (EEA), isobutylene isoprene rubber (IIR), ethylene / propylene copolymer rubber (EPR), etc. can be illustrated. Especially, ethylene / propylene copolymer rubber, and isobutylene isoprene rubber are suitable.

[0021] (C) As a plasticizer, a hydrocarbon system plasticizer is raised, for example. A liquid paraffin, polybutene, liquid polybutadiene, etc. can be illustrated as a hydrocarbon system plasticizer being concrete. moreover, various kinds, such as phthalic ester, an aliphatic series dibasic acid ester, glycol ester, phosphoric ester, and an epoxy system plasticizer, -- a well-known plasticizer is raised.

[0022] hot melt -- a sealing material -- a presentation -- a ratio -- thermoplastic elastomer -- (-- A --) -- containing -- the base -- a polymer -- 100 -- weight -- the section -- receiving -- -- usually -- said -- a tackifier -- (-- B --) -- 20 - 150 -- weight -- the section -- extent -- and -- a plasticizer -- (-- C --) -- five - 100 -- weight -- the section -- extent -- ** -- carrying out -- a thing -- being desirable . The good hot melt sealing material of balance with an adhesive property and detachability is obtained by this combination.

[0023] When under 20 weight sections of the loadings of said tackifier (B) are not enough as adhesive strength and they blend exceeding the 150 weight sections to the base polymer 100 weight section conversely, adhesive strength becomes strong too much and there is an inclination for a sealing material's detachability to get worse. As for the loadings of this viewpoint to said tackifier (B), it is more desirable to carry out to more than 25 weight sections and 130 or

less.

[0024] Under 5 weight sections of adhesive strength are not enough as the loadings of said plasticizer (C) to the base polymer 100 weight section, and there is an inclination for the regurgitation workability by the rise of melt viscosity to get worse. Conversely, when it blends exceeding the 100 weight sections, heat-resistant flow temperature falls and there is an inclination for a sealing material's detachability to get worse. As for the loadings of this viewpoint to a plasticizer (C), it is more desirable to carry out to more than 5 weight sections and 90 or less.

[0025] Although the hot melt sealing material of this invention contains the base polymer, tackifier (B), and plasticizer (C) containing thermoplastic elastomer (A) as aforementioned, he may add waxes, an inorganic bulking agent, a stabilizer, etc. in the range which does not deviate from the purpose of this invention further.

[0026] As waxes, paraffin wax, a micro crystallin wax, the Phi Shah Tropsch wax, low molecular weight polyethylene, low molecular weight polyethylene, low molecular weight polypropylene, bitter taste TAKUCHIKU polypropylene, these denaturation objects, etc. are used suitably. As an inorganic bulking agent, a calcium carbonate, a barium sulfate, talc, clay, titanium oxide, carbon black, etc. are used suitably. As a stabilizer, a hindered phenol system antioxidant, a hindered amine system antioxidant, various kinds of thermostabilizers, a weathering stabilizer, etc. are illustrated.

[0027] Although a hot melt sealing material is prepared by the above blending ratio of coal, as for the content of thermoplastic elastomer (A), it is desirable among a hot melt sealing material to blend each component so that it may become 15 - 70% of the weight. When the content of thermoplastic elastomer (A) decreases, since a sealing material's detachability is not fully desirable in respect of lightfastness, as for the content of thermoplastic elastomer (A), it is more desirable among a hot melt sealing material to consider as 40 % of the weight or more.

Conversely, when the content of thermoplastic elastomer (A) increases, an adhesive property is inadequate and, also as for the inclination for regurgitation workability to get worse by the rise of melt viscosity, it is more desirable [the content of thermoplastic elastomer (A)] among [from a certain thing] a hot melt sealing material to consider as 60 or less % of the weight.

[0028] Preparation of such a hot melt sealing material of this invention adjusts melt viscosity for the aforementioned (A) component, the (B) component and the (C) component, and the arbitration component blended if needed according to a component using equipments, such as a kneader. Usually, it is carried out by carrying out melting mixing at the temperature of about 180-240 degrees C.

[0029] In addition, there is especially no limitation in this invention's method of application and adhesion approach to various base materials of a hot melt sealing material, and various kinds are possible for the same approach as the usual hot melt constituent. For example, it is carried out by the approach using a hot melt applicator etc. A hot melt applicator consists of a hot melt fusion zone, the pump feeding section, a hose, and a gun. A tank type, a pail can type, or the drum type of a fusion zone is common. Moreover, as for the feeding section, the piston type, the gear pump type, the extrusion type, etc. are known. Furthermore, although a hose has the structure and the property that fixed temperature can be held, it is used, and a gun is an automatic gun or a hand gun.

[0030]

[Effect of the Invention] According to this invention, the tackifier for hot melt sealing materials used for the hot melt sealing material which was excellent in the lightfastness which becomes as a base polymer in thermoplastic elastomer, and the hot melt sealing material concerned can be offered. The hot melt sealing material of this invention is useful as a hot melt sealing material used for the part where the frequency exposed especially from the thing to excel in lightfastness is high. Moreover, the hot melt sealing material of this invention has the good balance of an adhesive property and detachability. That is, a good adhesive property is shown to various ingredients, such as polyolefine system resin, ABS system resin, acrylic resin, polycarbonate resin, and glass. Since the base material and base material which had been joined by compulsive exfoliation can be separated easily on the other hand and the sealing material concerned can

moreover be easily removed from a base material, recycle of a base material is possible.

[0031]

[Example] Although an example and the example of a comparison are raised to below and this invention is explained to it, this invention is not restricted to each [these] example.

[0032] Example 1 (manufacture of a tackifier)

By distilling C9 usual fraction (polymerization nature monomer presentation: vinyltoluene 37% and indene 35%, other 28%) obtained by cracking of naphtha, the polymerization of the purification C9 fraction (polymerization nature monomer presentation: vinyltoluene 58% and indene 9%, other 33%) which decreased the high-boiling point component was carried out by cationic polymerization, and special C9 system hydrocarbon resin (100 degrees C of softening temperatures, number average molecular weight 700) was obtained. The special C9 system hydrocarbon resin 100 section and the nickel diatom earth catalyst (product made from "N-113" JGC Chemistry) 2.0 section which were obtained were taught to the autoclave, and the hydrogenation reaction was performed to the bottom of 200kg/cm² of hydrogen pressure, the reaction temperature of 270 degrees C, and the condition of reaction-time 5 hours. The obtained resin was dissolved in the cyclohexane 300 section after reaction termination, and filtration removed the catalyst. Then, the filtrate and anti-oxidant ("IRUGA NOx 1010", Ciba-Geigy Japan make) 0.35 section was put into the separable flask of 1 liter capacity with which an impeller, the reflux capacitor, the thermometer, the temperature controller, and the pressure display meter were attached, a temperature up and reduced pressure of were gradually done to 200 degrees C and 20torr, the solvent was removed, and the hydrogenation C9 system hydrocarbon resin (tackifier a) 98 section of the quantity average molecular weight 790, 102.5 degrees C of softening temperatures, and 94% of rates of hydrogenation of a ring was obtained. The physical properties of the obtained tackifier a are shown in Table 1.

[0033] In addition, the rate of hydrogenation is raw material resin and obtained hydrogenation resin. Based on the following formulas, it computed from H-spectrum area of the ring which appears near 7 ppm of ¹H-NMR. Rate = of hydrogenation $[1 - (\text{spectrum area of spectrum area} / \text{raw material resin of hydrogenation resin})] \times 100 (\%)$. Moreover, softening temperature is JIS. It is based on the ring and ball method of K 2531.

[0034] Example 2 (manufacture of a tackifier)

In the example 1, hydrogenation conditions were changed as shown in Table 1, and also Tackifier b was manufactured like the example 1. The physical properties of the obtained tackifier b are shown in Table 1.

[0035] The example 1 (manufacture of a tackifier) of a comparison

It is cationic polymerization as it is about C9 usual fraction (polymerization nature monomer presentation: vinyltoluene 37% and indene 35%, other 28%) obtained by cracking of naphtha instead of special C9 system hydrocarbon resin in the example 1. Using C9 system hydrocarbon resin (120 degrees C of softening temperatures, number average molecular weight 740) which carried out the polymerization, hydrogenation conditions were changed, as shown in Table 1, and also the tackifier c of 95% of rates of hydrogenation of a ring was manufactured like the example 1. The physical properties of the obtained tackifier c are shown in Table 1.

[0036] The example 2 (manufacture of a tackifier) of a comparison

In the example 1 of a comparison, hydrogenation conditions were changed as shown in Table 1, and also the tackifier d of 70% of rates of hydrogenation of a ring was manufactured like the example 1 of a comparison. The physical properties of the obtained tackifier d are shown in Table 1.

[0037]

[Table 1]

	粘着付与剤	原料樹脂	水素化条件				粘着付与剤の物性		
			触媒量 (%)	温度 (°C)	水素圧 (kg/cm ²)	時間	軟化点 (°C)	数平均分子量	芳香環の水素化率 (%)
実施例 1	a	*1	2.0	270	200	5	102	790	94
実施例 2	b	*1	0.7	275	200	5	98	780	85
比較例 1	c	*2	1.2	300	200	5	100	700	95
比較例 2	d	*2	0.6	290	200	2	101	720	70

[0038] Front Naka and *1 show special C9 system hydrocarbon resin (polymerization nature monomer presentation: vinyltoluene 58% and indene 9%, other 33%, 100 degrees C of softening temperatures, number average molecular weight 700), and *2 show (polymerization nature monomer presentation: vinyltoluene 37% and indene 35%, other 28%, 120 degrees C of softening temperatures, and number average molecular weight 740) for C9 system hydrocarbon resin.

[0039] Examples 3-5, the examples 3-7 (preparation of a hot melt sealing material) of a comparison

After blending at thermoplastic elastomer (A), a tackifier (B) and a plasticizer (C), and a rate that shows other components in Table 2 further, it kneaded and pelletized at 220 degrees C using the biaxial kneading extruder, and the hot melt sealing material was obtained. [0040] The following evaluations were performed about the obtained hot melt sealing material. An evaluation result is shown in Table 2.

[0041] Viscosity: Viscosity was measured at 190 degrees C using the Brookfield viscometer.

[0042] Degree of hardness: It is JIS after cooling solidification. According to K6301 (Shore A), the value of 5 seconds after was measured at 20 degrees C.

[0043] Adhesive property (seal nature): The hot melt sealing material was applied to housing **** of the head lamp for automobiles at 200 degrees C using the extruder type [Nordson] hot melt applicator. The glass lens was stuck to the slot by pressure after radiational cooling, and it fixed, having covered the clip spring. It was immersed underwater and the existence of a leak was measured, after carrying out continuation lighting of this head lamp in a 50-degree C ambient atmosphere for 1000 hours. [0044] light-fast: -- a hot melt sealing material's pellet -- a xenon lamp -- 72-hour continuous irradiation -- carrying out -- yellowing -- viewing estimated the degree. [0045] In addition, the inside of Table 2, **:SEBS (made in [Shell Chemistry] trade name Clayton G-1652) and **SEPS (trade name SEPUTON 2002, Kuraray Make), ** hydrogenation petroleum resin (the trade name eye multiple independently targetable reentry vehicle P-100, product made from Idemitsu Petrochemistry), ** polybutene (trade name HV-100, product made from Nippon Oil Chemistry), ** wax (the trade name neo wax L, Yasuhara Chemical make), and ** anti-oxidant (trade name IRUGA NOx 1010, Ciba-Geigy Japan make) It is shown.

[0046]

[Table 2]

		実施例				比較例			
		3	4	5	6	3	4	5	6
シ リ ン グ 剤 の 組 成	A成分 ①	100	100	100	100	100	0	100	0
	②	0	0	0	0	0	100	0	100
	B成分 a	30	0	20	0	0	0	0	0
	b	0	30	0	0	0	0	0	0
	c	0	0	0	30	0	20	0	0
	d	0	0	0	0	30	0	0	0
	⑤	0	0	0	0	0	0	30	20
	C成分 ④	50	50	50	50	50	50	50	50
	その他 ⑤	55	55	0	55	55	0	55	0
	⑥	1	1	1	1	1	1	1	1
性 能 評 価	粘度 (cP)	82000	76000	26000	78000	74000	25000	77000	23000
	硬度	68	64	34	68	63	33	65	34
評 価	シール性(水漏れ)	無し	無し	無し	無し	無し	無し	無し	無し
	耐光性	無色	微黄色	無色	黄色	黄色	黄色	黄色	黄色

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TECHNICAL FIELD

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[0003] Since it excels in colorlessness and transparency, the sealing material which makes thermoplastic elastomer a base polymer also in said thermoplastic polymer is used as a clear sealing material of the part where importance is attached to colorlessness, such as junction on the lens and the housing body which are used for the tail lamp for cars, or a head lamp, and transparency. Moreover, since junction of a base material is performed by hot melt (thermofusion liquid) coating, compared with the sealing material of the type which joins completely the base material which makes a base polymer thermoplastic polymers, such as thermosetting polyurethane, and ethylene / vinyl acetate copolymer, a polyamide, exfoliation of a base material is comparatively easy for this clear sealing material, and also when performing the exchange and collection by type of a base material by type, he is desirable.

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EFFECT OF THE INVENTION

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MEANS

[Means for Solving the Problem] this invention persons came to complete header this invention for the ability of said purpose to be attained by using the hydride of the specific aromatic series system hydrocarbon resin shown below as a tackifier for hot melt sealing materials, as a result of repeating examination wholeheartedly that said technical problem should be solved.

[0008] Namely, this invention is a tackifier for hot melt sealing materials which comes to use the hydride of the aromatic series system hydrocarbon resin obtained by carrying out the polymerization of the polymerization nature monomer in C9 fraction. This polymerization nature monomer contains vinyltoluene and contains an indene at 20 or less % of the weight of a rate 50% of the weight or more. and to the tackifier for hot melt sealing materials and pan which are characterized by the rate of hydrogenation of the ring of aromatic series system hydrocarbon resin being 50% or more thermoplastic elastomer -- (--- A ---) -- containing -- becoming -- the base -- a polymer -- 100 -- weight -- the section -- receiving -- said -- hot melt -- a sealing material -- ** -- a tackifier -- (--- B ---) -- 20 - 150 -- weight -- the section -- and -- a plasticizer -- (--- C ---) -- five - 100 -- weight -- the section -- blending -- becoming -- hot melt -- C -- RUNGU -- material -- being related .

[0009]

[Embodiment of the Invention] The tackifier for hot melt sealing materials of this invention (B) is the hydride of the aromatic series system hydrocarbon resin obtained by carrying out the polymerization of the polymerization nature monomer in C9 fraction, this polymerization nature monomer contains vinyltoluene 50% of the weight or more, and contains an indene at 20 or less % of the weight of a rate, and the rate of hydrogenation of the ring of aromatic series system hydrocarbon resin is 50% or more of thing.

[0010] C9 fraction usually means what was obtained according to cracking of naphtha, and the compound of high-boiling in addition points, such as styrene, vinyltoluene, alpha methyl styrene, and indenenes, etc. is contained as a polymerization nature monomer in C9 fraction. C9 general fraction usually contains vinyltoluene and an indene about 40% of the weight, respectively, and about remaining 20 % of the weight consists of styrene etc. In this invention, by distilling such C9 fraction, the indenenes of C9 fraction, the compound of a high-boiling point, etc. are removed, and the vinyltoluene content in a polymerization nature monomer uses what was prepared so that the content of an indene might become 20 or less % of the weight at 50 % of the weight or more. It is 60 % of the weight or more more preferably 55% of the weight or more, it is so good that many, and preferably, the content of an indene is 10 or less % of the weight more preferably, and it is preferably good [few vinyltoluene contents] so that there are 15 or less % of the weight. When the content of the vinyltoluene in a polymerization nature monomer is less than 50%, or when the content of an indene exceeds 20 % of the weight, it becomes impossible to be satisfied with the hydride of the aromatic series system hydrocarbon resin obtained of lightfastness. In addition, aromatic series system hydrocarbon resin is obtained by carrying out the polymerization of this polymerization nature monomer by the usual cationic polymerization.

[0011] Said aromatic series system hydrocarbon resin is hydrogenated so that the rate of hydrogenation of a ring may become 50% or more further. Lightfastness cannot be satisfied when the rate of hydrogenation of a nucleus is less than 50%.

[0012] A hydrogenation reaction is performed by adjusting conditions suitably to the bottom of existence of a hydrogenation catalyst so that the rate of hydrogenation of said aromatic series system hydrocarbon resin may become said within the limits (the rate of hydrogenation of a ring is 50% or more).

[0013] As a hydrogenation catalyst, various kinds of things, such as metallic compounds, such as metals, such as nickel, palladium, platinum, cobalt, a rhodium, a ruthenium, and molybdenum, or these oxides, and a sulfide, can be used. This hydrogenation catalyst may be supported and used for support, such as an alumina with big surface area, a silica (diatom earth), carbon, and a titania, by porosity. It is desirable to use a nickel-diatom earth catalyst also in these catalysts in this invention from that it is easy to adjust the rate of hydrogenation to said within the limits or a costs side. 0.1- of the aromatic series system hydrocarbon resin whose amount of the catalyst used is raw material resin -- it is 0.1 - 2 % of the weight preferably about 3% of the weight.

[0014] A hydrogenation pressure is performed in the about two 30 - 300 kg/cm range, and the conditions of a hydrogenation reaction usually perform reaction temperature in about 150-300 degrees C. A hydrogenation pressure is 100 - 200 kg/cm² preferably, and reaction temperature is 200-280 degrees C. When a hydrogenation pressure does not fulfill 30 kg/cm², or when reaction temperature does not fulfill 150 degrees C, hydrogenation cannot progress easily, and when a hydrogenation pressure exceeds 300 kg/cm², or when reaction temperature exceeds 300 degrees C, there is an inclination for decomposition to take place and for softening temperature to fall. Moreover, reaction time is usually 2 - 7 hours preferably for about 1 to 7 hours. Said hydrogenation reaction is performed in the condition of having fused aromatic series system hydrocarbon resin, or having dissolved in the solvent. A cyclohexane, n-hexane, n-heptane, a decalin, etc. can be used as a solvent.

[0015] In addition, although the case where a batch process was adopted as a reaction format about the amount used and reaction time of a catalyst was explained, as a reaction format, circulation types (a fixed-bed type, fluidized bed, etc.) are also employable.

[0016] In this way, as for about 60-130 degrees C and number average molecular weight, 300 to about 2000 are [the softening temperature of the hydride of the obtained aromatic series system hydrocarbon resin] usually desirable.

[0017] The hot melt sealing material of this invention blends said tackifier for hot melt sealing materials (B) and plasticizer (C) with the base polymer containing thermoplastic elastomer (A).

[0018] As thermoplastic elastomer (A), the copolymer or block copolymer of styrene, such as styrene and methyl styrene, and conjugated dienes, such as a butadiene and an isoprene, is raised. Although especially the ratio of the weight ratio of styrene/conjugated dienes is not restricted, it is usually about 10 / 90 to 50/50.

[0019] As thermoplastic elastomer (A) being concrete, a styrene-butadiene copolymer, a styrene-isoprene copolymer, a styrene-isoprene-styrene triblock copolymer (SIS), a styrene-butadiene-styrene triblock copolymer (SBS), etc. can be illustrated, for example. Furthermore, as thermoplastic elastomer (A), what hydrogenated said copolymer or block copolymers, such as a hydride of a styrene-butadiene copolymer, a hydride of a styrene-isoprene copolymer, a hydride [(styrene/ethylene) - (butylene/styrene) copolymer] (SEBS) of SBS, and a hydride [(styrene/ethylene) - (propylene/styrene) copolymer] (SEPS) of SIS, and its denaturation object are raised. In addition, various kinds of denaturation objects, such as what especially the rate of hydrogenation of said copolymer or a block copolymer was not *(ed), and introduced polar groups, such as a carboxyl group and a hydroxyl group, into some of said copolymers or block copolymers as a denaturation object, are raised. As thermoplastic elastomer (A) of this this invention, the hydride and/or its denaturation objects of a styrene-conjugated diene system block copolymer, such as SEBS and SEPS, are desirable at the point of excelling in lightfastness.

[0020] Since the adhesion to adherend and the adhesive property at the time of heating are raised in addition to said thermoplastic elastomer (A), thermoplastic polymers other than said thermoplastic elastomer (A) can also be used for the base polymer of the hot melt sealing material of this invention. As such a thermoplastic polymer, ethylene / vinyl acetate copolymer (EVA), an ethylene ethyl acrylate copolymer (EEA), isobutylene isoprene rubber (IIR), ethylene /

propylene copolymer rubber (B), etc. can be illustrated. Especially, ethylene / propylene copolymer rubber, and isobutylene isoprene rubber are suitable.

[0021] (C) As a plasticizer, a hydrocarbon system plasticizer is raised, for example. A liquid paraffin, polybutene, liquid polybutadiene, etc. can be illustrated as a hydrocarbon system plasticizer being concrete. moreover, various kinds, such as phthalic ester, an aliphatic series dibasic acid ester, glycol ester, phosphoric ester, and an epoxy system plasticizer, -- a well-known plasticizer is raised.

[0022] hot melt -- a sealing material -- a presentation -- a ratio -- thermoplastic elastomer -- (A) -- containing -- the base -- a polymer -- 100 -- weight -- the section -- receiving -- usually -- said -- a tackifier -- (B) -- 20 - 150 -- weight -- the section -- extent -- and -- a plasticizer -- (C) -- five - 100 -- weight -- the section -- extent -- ** -- carrying out -- a thing -- being desirable . The good hot melt sealing material of balance with an adhesive property and detachability is obtained by this combination.

[0023] When under 20 weight sections of the loadings of said tackifier (B) are not enough as adhesive strength and they blend exceeding the 150 weight sections to the base polymer 100 weight section conversely, adhesive strength becomes strong too much and there is an inclination for a sealing material's detachability to get worse. As for the loadings of this viewpoint to said tackifier (B), it is more desirable to carry out to more than 25 weight sections and 130 or less.

[0024] Under 5 weight sections of adhesive strength are not enough as the loadings of said plasticizer (C) to the base polymer 100 weight section, and there is an inclination for the regurgitation workability by the rise of melt viscosity to get worse. Conversely, when it blends exceeding the 100 weight sections, heat-resistant flow temperature falls and there is an inclination for a sealing material's detachability to get worse. As for the loadings of this viewpoint to a plasticizer (C), it is more desirable to carry out to more than 5 weight sections and 90 or less.

[0025] Although the hot melt sealing material of this invention contains the base polymer, tackifier (B), and plasticizer (C) containing thermoplastic elastomer (A) as aforementioned, he may add waxes, an inorganic bulking agent, a stabilizer, etc. in the range which does not deviate from the purpose of this invention further.

[0026] As waxes, paraffin wax, a micro crystallin wax, the Phi Shah Tropsch wax, low molecular weight polyethylene, low molecular weight polyethylene, low molecular weight polypropylene, bitter taste TAKUCHIKU polypropylene, these denaturation objects, etc. are used suitably. As an inorganic bulking agent, a calcium carbonate, a barium sulfate, talc, clay, titanium oxide, carbon black, etc. are used suitably. As a stabilizer, a hindered phenol system antioxidant, a hindered amine system antioxidant, various kinds of thermostabilizers, a weathering stabilizer, etc. are illustrated.

[0027] Although a hot melt sealing material is prepared by the above blending ratio of coal, as for the content of thermoplastic elastomer (A), it is desirable among a hot melt sealing material to blend each component so that it may become 15 - 70% of the weight. When the content of thermoplastic elastomer (A) decreases, since a sealing material's detachability is not fully desirable in respect of lightfastness, as for the content of thermoplastic elastomer (A), it is more desirable among a hot melt sealing material to consider as 40 % of the weight or more. Conversely, when the content of thermoplastic elastomer (A) increases, an adhesive property is inadequate and, also as for the inclination for regurgitation workability to get worse by the rise of melt viscosity, it is more desirable [the content of thermoplastic elastomer (A)] among [from a certain thing] a hot melt sealing material to consider as 60 or less % of the weight.

[0028] Preparation of such a hot melt sealing material of this invention adjusts melt viscosity for the aforementioned (A) component, the (B) component and the (C) component, and the arbitration component blended if needed according to a component using equipments, such as a kneader. Usually, it is carried out by carrying out melting mixing at the temperature of about 180-240 degrees C.

[0029] In addition, there is especially no limitation in this invention's method of application and adhesion approach to various base materials of a hot melt sealing material, and various kinds are

possible for the same approach as the usual hot melt constituent. For example, it is carried out by the approach using a hot melt applicator etc. A hot melt applicator consists of a hot melt fusion zone, the pump feeding section, a hose, and a gun. A tank type, a pail can type, or the drum type of a fusion zone is common. Moreover, as for the feeding section, the piston type, the gear pump type, the extrusion type, etc. are known. Furthermore, although a hose has the structure and the property that fixed temperature can be held, it is used, and a gun is an automatic gun or a hand gun.

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EXAMPLE

[Example] Although an example and the example of a comparison are raised to below and this invention is explained to it, this invention is not restricted to each [these] example.

[0032] Example 1 (manufacture of a tackifier)

By distilling C9 usual fraction (polymerization nature monomer presentation: vinyltoluene 37% and indene 35%, other 28%) obtained by cracking of naphtha, the polymerization of the purification C9 fraction (polymerization nature monomer presentation: vinyltoluene 58% and indene 9%, other 33%) which decreased the high-boiling point component was carried out by cationic polymerization, and special C9 system hydrocarbon resin (100 degrees C of softening temperatures, number average molecular weight 700) was obtained. The special C9 system hydrocarbon resin 100 section and the nickel diatom earth catalyst (product made from "N-113" JGC Chemistry) 2.0 section which were obtained were taught to the autoclave, and the hydrogenation reaction was performed to the bottom of 200kg/cm² of hydrogen pressure, the reaction temperature of 270 degrees C, and the condition of reaction-time 5 hours. The obtained resin was dissolved in the cyclohexane 300 section after reaction termination, and filtration removed the catalyst. Then, the filtrate and anti-oxidant ("IRUGA NOx 1010", Ciba-Geigy Japan make) 0.35 section was put into the separable flask of 1 liter capacity with which an impeller, the reflux capacitor, the thermometer, the temperature controller, and the pressure display meter were attached, a temperature up and reduced pressure of were gradually done to 200 degrees C and 20torr, the solvent was removed, and the hydrogenation C9 system hydrocarbon resin (tackifier a) 98 section of the quantity average molecular weight 790, 102.5 degrees C of softening temperatures, and 94% of rates of hydrogenation of a ring was obtained. The physical properties of the obtained tackifier a are shown in Table 1.

[0033] In addition, the rate of hydrogenation is raw material resin and obtained hydrogenation resin. Based on the following formulas, it computed from H-spectrum area of the ring which appears near 7 ppm of ¹H-NMR. Rate = of hydrogenation $\{1 - (\text{spectrum area of spectrum area} / \text{raw material resin of hydrogenation resin})\} \times 100$ (%). Moreover, softening temperature is JIS. K It is based on the ring and ball method of 2531.

[0034] Example 2 (manufacture of a tackifier)

In the example 1, hydrogenation conditions were changed as shown in Table 1, and also Tackifier b was manufactured like the example 1. The physical properties of the obtained tackifier b are shown in Table 1.

[0035] The example 1 (manufacture of a tackifier) of a comparison

It is cationic polymerization as it is about C9 usual fraction (polymerization nature monomer presentation: vinyltoluene 37% and indene 35%, other 28%) obtained by cracking of naphtha instead of special C9 system hydrocarbon resin in the example 1. Using C9 system hydrocarbon resin (120 degrees C of softening temperatures, number average molecular weight 740) which carried out the polymerization, hydrogenation conditions were changed, as shown in Table 1, and also the tackifier c of 95% of rates of hydrogenation of a ring was manufactured like the example 1. The physical properties of the obtained tackifier c are shown in Table 1.

[0036] The example 2 (manufacture of a tackifier) of a comparison

In the example 1 of a comparison, hydrogenation conditions were changed as shown in Table 1,

and also the tackifier d of 70% of rates of hydrogenation of a ring was manufactured like the example 1 of a comparison. The physical properties of the obtained tackifier d are shown in Table 1.

[0037]

[Table 1]

	粘着付与剤	原料樹脂	水素化条件				粘着付与剤の物性		
			触媒量 (%)	温度 (°C)	水素圧 (kg/cm ²)	時間	軟化点 (°C)	数平均分子量	芳香環の水素化率 (%)
実施例 1	a	*1	2.0	270	200	5	102	790	94
実施例 2	b	*1	0.7	275	200	5	98	780	85
比較例 1	c	*2	1.2	300	200	5	100	700	95
比較例 2	d	*2	0.6	290	200	2	101	720	70

[0038] Front Naka and *1 show special C9 system hydrocarbon resin (polymerization nature monomer presentation: vinyltoluene 58% and indene 9%, other 33%, 100 degrees C of softening temperatures, number average molecular weight 700), and *2 show (polymerization nature monomer presentation: vinyltoluene 37% and indene 35%, other 28%, 120 degrees C of softening temperatures, and number average molecular weight 740) for C9 system hydrocarbon resin.

[0039] Examples 3-5, the examples 3-7 (preparation of a hot melt sealing material) of a comparison

After blending at thermoplastic elastomer (A), a tackifier (B) and a plasticizer (C), and a rate that shows other components in Table 2 further, it kneaded and pelletized at 220 degrees C using the biaxial kneading extruder, and the hot melt sealing material was obtained. [0040] The following evaluations were performed about the obtained hot melt sealing material. An evaluation result is shown in Table 2.

[0041] Viscosity: Viscosity was measured at 190 degrees C using the Brookfield viscometer.

[0042] Degree of hardness: It is JIS after cooling solidification. According to K6301 (Shore A), the value of 5 seconds after was measured at 20 degrees C.

[0043] Adhesive property (seal nature): The hot melt sealing material was applied to housing **** of the head lamp for automobiles at 200 degrees C using the extruder type [Nordson] hot melt applicator. The glass lens was stuck to the slot by pressure after radiational cooling, and it fixed, having covered the clip spring. It was immersed underwater and the existence of a leak was measured, after carrying out continuation lighting of this head lamp in a 50-degree C ambient atmosphere for 1000 hours. [0044] light-fast: -- a hot melt sealing material's pellet -- a xenon lamp -- 72-hour continuous irradiation -- carrying out -- yellowing -- viewing estimated the degree. [0045] In addition, the inside of Table 2, **:SEBS (made in [Shell Chemistry] trade name Clayton G-1652) and **SEPS (trade name SEPUTON 2002, Kuraray Make), ** hydrogenation petroleum resin (the trade name eye multiple independently targetable reentry vehicle P-100, product made from Idemitsu Petrochemistry), ** polybutene (trade name HV-100, product made from Nippon Oil Chemistry), ** wax (the trade name neo wax L, Yasuhara Chemical make), and ** anti-oxidant (trade name IRUGA NOx 1010, Ciba-Geigy Japan make) It is shown.

[0046]

[Table 2]

		実施例			比較例				
		3	4	5	3	4	5	6	7
シ- リン グ 剤 の 組 成	A成分 ①	100	100	100	100	100	0	100	0
	②	0	0	0	0	0	100	0	100
	B成分 a	30	0	20	0	0	0	0	0
	b	0	30	0	0	0	0	0	0
	c	0	0	0	30	0	20	0	0
	d	0	0	0	0	30	0	0	0
	⑤	0	0	0	0	0	0	30	20
	C成分 ④	50	50	50	50	50	50	50	50
	その他 ⑥	55	55	0	55	55	0	55	0
	⑦	1	1	1	1	1	1	1	1
性	粘度 (cP)	82000	76000	26000	78000	74000	25000	77000	23000
能	硬度	6.8	6.4	3.4	6.8	6.8	8.8	6.5	8.4
評	シール性 (水漏れ)	無し	無し	無し	無し	無し	無し	無し	無し
価	耐光性	無色	微黄色	無色	黄色	黄色	黄色	黄色	黄色

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